

# Simulations of prompt many-body ionization in a frozen Rydberg gas

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The results of a theoretical investigation of prompt many-body ionization are reported. Our calculations address an experiment that reported ionization in Rydberg gases for densities two orders of magnitude less than expected from ionization between pairs of atoms. The authors argued that the results were due to the simultaneous interaction between many atoms. We performed classical calculations for many interacting Rydberg atoms with the ions fixed in space and have found that the many atom interaction does allow ionization at lower densities than estimates from two atom interactions. However, we found that the density fluctuations in a gas play a larger role. These two effects are an order of magnitude too small to account for the experimental results suggesting at least one other mechanism strongly affects ionization.

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## I. INTRODUCTION

Most low density gases (e.g.  $10^{10} \text{ cm}^{-3}$ ) consist of nearly independent atoms or molecules that interact through random binary collisions. Gases consisting of Rydberg atoms can violate this picture strongly. Even though the sizes of the atoms are small compared to the spacing between the atoms, the large dipole moments that can be formed allow for a large interaction between atoms. There have been many recent experiments where the atoms have been separated by 10's of  $\mu\text{m}$ , but still showed strong interaction. The strong interaction between the atoms and the controllability inherent in exciting specific states has led to the possibility of using Rydberg gases as examples of many-body systems.

A recent experiment[1] found that ionization in a frozen Rydberg gas occurred at much lower densities than expected from calculations of pair-wise interactions.[2] In this calculation, it was found that 90% of the trajectories led to ionization when a pair of atoms were at a separation of  $\sim 2.1 \times 2n^2 a_0$ . In Ref. [1], they used

$$\rho = \frac{1}{(4\pi/3)(4n^2 a_0)^3} = \frac{3}{256\pi n^6 a_0^3} \quad (1)$$

as the reference density; this density corresponds to one atom within a sphere of radius  $4n^2 a_0$ . For a Rb 45d state,  $n \simeq 43.7$  and the reference density is  $\sim 4 \times 10^{12} \text{ cm}^{-3}$ . In their experiment, they excite the atoms to the 45d state, wait 10 ns, and then ramp an electric field to measure the ions and atoms. The details of the measurement means that they measure the ionization approximately “100 ns after laser excitation of the frozen Rydberg gas.” The moniker “frozen Rydberg gas” is applicable because the Rb atoms have a temperature of 300  $\mu\text{K}$  giving an RMS

speed of  $\sim 0.3 \text{ m/s}$ . Thus, during 100 ns, the atoms move  $\sim 30 \text{ nm}$  which is much smaller than the size of the atoms,  $\sim 200 \text{ nm}$ , and is much, much smaller than the spacing between atoms,  $\sim 1/\rho_{exp}^{1/3} \sim 3000 \text{ nm}$ . Because the atoms travel such a small distance, they don't ionize through collisional processes.

At a density of  $5 \times 10^{10} \text{ cm}^{-3}$ , they measured substantial ionization at early times. The surprisingly large amount of ionization was attributed to many-body interactions since the ionization occurs at densities roughly 2 orders of magnitude less than the base density needed for ionization between pairs of atoms, Eq. (1). There have been several other experimental studies of ionization in a Rydberg gas or the conversion of the Rydberg gas to a plasma (e.g. see Refs. [3–12]). However, these studies are fundamentally different from Ref. [1] in that the time scale of the ionization is much longer and the atoms move a substantial fraction of their spacing. There was a quantum calculation of the autoionization from pairs of Rydberg atoms[13], but the authors found that this quantum effect was negligible for the situation of Ref. [1].

In order to test the idea of many-body ionization, we performed classical trajectory Monte Carlo calculations of many interacting Rydberg atoms. Since the atoms are in highly excited states and the physics involves substantial averaging, we expect that classical calculations will provide a good approximation to the actual quantum physics. The advantage of the classical calculation is that we can include all of the electron-electron interactions without approximation. Thus, the ionization process will be properly represented even if it requires the interaction between many widely spaced atoms.

There is an important difference between classical and quantum calculations of ionization for a pair of Rydberg atoms. In the quantum calculation, there is a nonzero matrix element from the dipole-dipole interaction which can cause one electron to go to a more deeply bound state and the other electron to be ionized. Since the

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ionized electron is in the continuum, this is the analog of autoionization.[13] Thus, for large separations,  $R$ , the quantum decay rate decreases like  $1/R^6$  since the matrix element is proportional to  $1/R^3$ . In contrast, the classical ionization probability becomes 0 outside of a separation not much larger than  $3.5\times$  the atom size. The classical ionization probability drops exactly to 0 because as one electron gains energy (thus decreasing its Rydberg frequency) the other electron loses energy (thus increasing its Rydberg frequency). This is analogous to driving a pendulum exactly on resonance for small angle oscillations (if the coupling is weak, the oscillator gains energy until the oscillation frequency changes enough to put it out of phase with the drive). The energy of each atom oscillates around the average energy with a spread that decreases as the separation of atoms increase. This difference in ionization at large separation is not important for the calculations in this paper because we are interested in delimiting the densities where the ionization is fast from those where it is slow.

In a real gas, the atoms have a random spacing and the distribution of spacing affects the amount of ionization. In order to control for this, we performed calculations for the unphysical situation where the atoms have a fixed spacing. We performed calculations for particles on a line, on a square array, and on a cubic grid. For these cases, any ionization for separations larger than the maximum ionization for a pair of atoms will necessarily be due to many-body interactions. We compare these results to calculations for atoms randomly distributed in space. We found that the character of ionization substantially changed when going to a random distribution. In our calculations, the density fluctuations play a larger role in ionization than the many-body interactions. However, our results could not reproduce the experimental results which suggests there is at least one other important mechanism for ionization in a dense Rydberg gas.

## II. NUMERICAL METHOD

Our calculations are purely classical where the electrons obey Newton's equations and the nuclei are fixed in space. We solve the coupled first order equations in  $\vec{v}, \vec{r}$  using an adaptive step size Runge-Kutta algorithm similar to that in Ref. [14]. The main change is in how we scale the variables with all of the velocity components of the  $i$ -th electron being scaled by the speed of the  $i$ -th electron and the position components being scaled by the distance to the closest nucleus. For each set of initial conditions, we checked the change in total energy at the final time. If the energy drifted by more than 0.1%, the trajectory was rerun with the same initial conditions but the error scale decreased by an order of magnitude. The process was repeated until the energy drift was less than our set value. We tested that our results were converged with respect to the setting of our accuracy parameter.

We defined ionization to be when any electron reaches

a distance more than 100 atom spacings from the central position of the many atom system. We chose 2,000 Rydberg periods for the final time of the calculation. This is long enough that most of the trajectories that lead to ionization will have an electron reach the final distance. But it is not so long that we waste computer time solving trajectories that will never lead to ionization.

We used a perfect Coulomb force for the electron-electron interactions, but a soft-core force for the electron-nuclei interactions. The potential energy between an electron and nucleus was proportional to  $-1/\sqrt{r^2 + b^2}$  where  $r$  is the distance between the electron and nucleus and  $b$  is a constant. Because of a non-zero  $b$ , the potential is not singular and the force does not diverge as  $r \rightarrow 0$ . Defining a principal quantum number from the electron's launch energy using  $-13.6 \text{ eV}/n^2$ , we chose  $b = 2(n/20)^2 a_0$  with  $a_0$  the Bohr radius; this gives a screening length  $b$  that is  $\sim 1/400$  the size of the atom. We found that choosing  $b = a_0$  gave similar results at the price of longer calculations. In all of our calculations, one electron was launched from near each nucleus. This simulates the photo-excitation step. For all calculations, we launched the electrons with  $n = 60$ . Because this classical system scales with  $n$  and we present all our results in terms of ratios, the actual value of  $n$  chosen is not relevant. At small enough  $n$ , quantum effects will become important but that is beyond the scope of this paper.

For the grid calculations, the nuclei were exactly on a grid of points in one, two or three dimensions. For the random calculations, the  $x$ -,  $y$ -, and  $z$ -positions were chosen from a flat distribution between 0 and  $L$ . In Figs. 4-6, the separation  $D$  is defined to be  $L$  divided by the number of atoms for one dimension,  $L$  divided by the square root of the number of atoms for two dimensions, and  $L$  divided by the cube root of the number of atoms for three dimensions.

In an experiment, the electrons are not simultaneously excited to the Rydberg state but randomly absorb photons proportional to the time dependent laser intensity. This duration will depend on the specific laser and the effect will depend on the ratio of the duration to the classical Rydberg period of the state being excited. Clearly, we don't want to launch all electrons at the same time because they will initially have the same phase in the classical orbit. We did calculations where the time of each electron's launch was random with a flat distribution between 0 and 1 Rydberg period or with a flat distribution between 0 and 100 Rydberg periods. We found that the results quantitatively depended on how we launched the electrons but the qualitative results we were after did not depend on how the random times were chosen.

Finally, we chose the initial position of each electron to be randomly on a sphere of radius  $r_0 = 2n^2 a_0/100$  centered on its nucleus. The speed of the electron is determined by its energy  $-13.6 \text{ eV}/n^2$ . The direction of the velocity was chosen to be perpendicular to the radius making  $r_0$  the perigee of the orbit for an isolated atom. The direction of the velocity was random in the plane

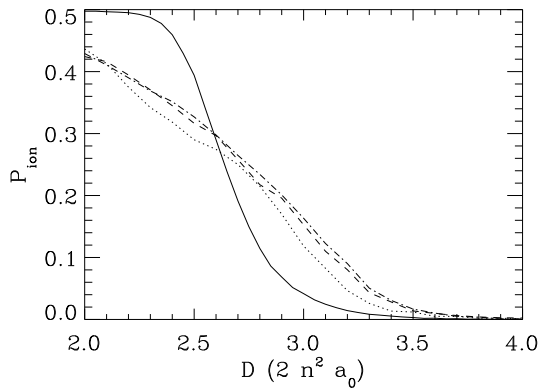


FIG. 1: The probability for an atom to be ionized as a function of the atom separation for atoms in a line. The different line-types correspond to different number of atoms: 2 atoms (solid), 4 atoms (dotted), 8 atoms (dashed), and 16 atoms (dash-dot).

perpendicular to  $\hat{r}$  with  $\vec{v} = (\hat{\theta} \cos(\alpha) + \hat{\phi} \sin(\alpha))v$  where  $\alpha$  was from a flat distribution between 0 and  $2\pi$ .

Of all the choices for initial conditions, our results most strongly depended on our choice of  $r_0$ . If instead, we chose a microcanonical ensemble as in Ref. [2] our ionization curves shift to smaller atom separation by  $\sim 10\%$ . As with other choices described above, the general trends and conclusions do not depend on  $r_0$ .

### III. RESULTS

We performed two styles of calculations to try to cleanly show the effect of many-body ionization. In one set of calculations, we have the nuclei on equally spaced points in one dimension, on a square lattice in two dimensions, or on a cubic lattice in three dimensions. For these cases, there is a limit on the atoms' smallest separation and, thus, any increase over independent pairs is an indication of many-body ionization. As might be expected, the many-body effect is more apparent with increasing dimension.

The other set of calculations is to randomly place atoms on a line, within a square, or within a cube. Besides many-body ionization, now there can be pairs of atoms that are randomly close enough to quickly ionize. This second effect leads to substantially more ionization compared to a grid of nuclei.

All plots show the probability for ionization as a function of atom spacing. The probability for ionization is the same as the fraction of atoms that ionized averaged over all of the calculations with different initial conditions.

#### A. Atoms on a Regular Grid

In Fig. 1, we plot the fraction of atoms ionized versus the separation of atoms for different number of atoms. The solid line is for a pair of atoms. We can compare this result to that reported in Ref. [2] by multiplying the curve in Fig. 1 by a factor of two because the fraction of trajectories leading to ionization is just two times the fraction of atoms ionized for calculations with a pair of atoms. The present result slightly differs from that reported in Ref. [2] in that 90% or more of the trajectories lead to ionization for scaled separations of 2.3 in Fig. 1 while the value was 2.1 in Ref. [2]. This difference is due to the choice made for the initial electron conditions as discussed in Sec. II.

The results for the two atom case have the simplest explanation. As the separation decreases, the probability that at least one atom will ionize rises to nearly one. There is a rapid drop in ionization probability between 2.5 and 3.0 which reflects the decreased coupling between the atoms and the destruction of the resonance condition as energy is exchanged between atoms. As one atom gains energy, its Rydberg period increases while the Rydberg period for the atom that loses energy decreases. When atoms are widely separated, this destruction of the resonance condition prevents ionization.

There is a large change in the ionization probability when going from 2 to 4 atoms. For small separation, there is a *decrease* in the fraction of atoms ionized. This is because the atoms might not ionize in ordered pairs. For example, atoms 2 and 3 might quickly ionize in the 4 atom case leaving atoms 1 and 4 far away from atoms that they can strongly interact with. This only needs to occur in approximately 20% of the runs to obtain the effect seen in Fig. 1. For larger separation, there is an *increase* in the ionization fraction due to many-body ionization as discussed in Ref. [1]. Compared to the two atom case, the atoms have more near neighbors. There is a greater chance for exchanging energy. Also, the destruction of the resonance condition for the two atom case does not necessarily hold for more atoms. For example, atom 2 could gain energy from atom 1 while atom 3 gains energy from atom 4; this will leave atoms 2 and 3 in (near) resonance and they can continue the exchange of energy until one of them ionizes.

Note that the 8-atom and 16-atom results are nearly identical. This shows that the one-dimensional case quickly converges with respect to the number of atoms. There are only two atoms at the edge of the grid and, thus, the effect of finite atom number is small.

We also calculated the fraction of configurations that led to *at least one* ionization. This is not directly related to an experimental observable, but lends itself to an easy test of many-body ionization. As an example, the 4 atom case has 3 pairs of atoms with a separation  $R$ . The probability for at least one ionization if each pair independently ionizes is one minus the probability for all pairs to *not* ionize. Even for this case, we found that there is

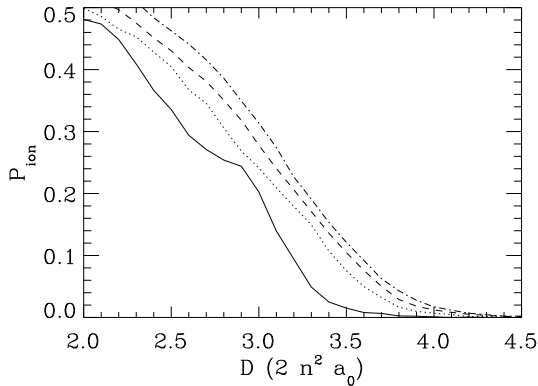


FIG. 2: The probability for ionization as a function of the atom separation for atoms in a square lattice. The different line-types correspond to different number of atoms:  $2^2$  atoms (solid),  $3^2$  atoms (dotted),  $4^2$  atoms (dashed), and  $5^2$  atoms (dash-dot).

more ionization than can be accounted for simply by the increased number of pairs of atoms. Thus, there must be some cooperativity in the ionization process which can be counted as many-body ionization. However, we found that a large part of the increase is simply due to the increase in number of atom pairs.

Figure 2 shows the fraction of ionized atoms in a square array versus their separation for different number of atoms. Note there is a slightly larger  $x$ -range in Fig. 2 compared to Fig. 1. Unlike the one-dimensional case, the fraction of atoms ionized increases with the number of atoms for the full range of separations shown. Also, there does not seem to be convergence with respect to the number of atoms in a simulation. This is partly due to the larger fraction of atoms on the surface of the grid. Even the case with  $5^2$  atoms has more surface atoms than interior atoms: 4 corner atoms, 12 edge atoms, and 9 interior atoms.

As expected, there is more ionization for the two dimensional case compared to one dimension because each atom has more neighbors that are close. This leads to a net stronger interaction and, thus, a larger fraction of atoms ionize. To quantify the increase of ionization, we note that approximately 10% of the atoms ionize for the one dimensional case with 16 atoms at a separation  $D = 3.2$  compared to the two dimensional case with 25 atoms at a separation  $D = 3.6$ . Due to the lack of convergence with respect to atom number, we do not have a firm prediction of the large atom limit of the fraction of ionized atoms.

Figure 3 shows the fraction of ionized atoms in a cubic array versus their separation for different number of atoms. Note there is a slightly larger  $x$ -range in Fig. 3 compared to Figs. 1 and 2. We only have two examples of a cubic array because the number of atoms increases very rapidly in three dimensions and the computer time

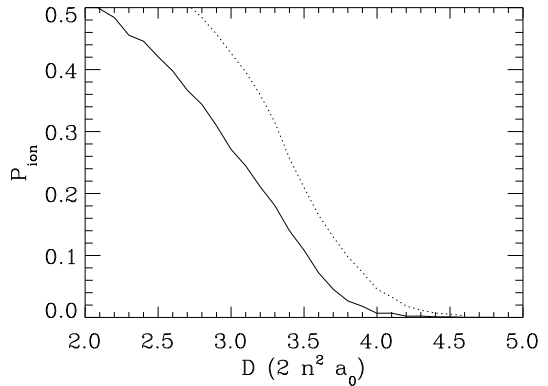


FIG. 3: The probability for ionization as a function of the atom separation for atoms in a cubic array. The different line-types correspond to different number of atoms:  $2^3$  atoms (solid) and  $3^3$  atoms (dotted),

scales approximately with the third power of the number of atoms. Because there is such a large change between the  $2^3$  and  $3^3$  cases, we can not predict how large an increase in ionization would be present for a large number of atoms.

Even though the three dimensional case is not converged, it's clear that there is more ionization than in the one and two dimensional cases. Approximately 10% of the atoms ionize for the three dimensional case with 27 atoms at a separation  $D = 3.8$ . If we compare to the one dimensional case with 2 atoms ( $D = 3.0$ ), the increase in separation does not appear to be very large (i.e. approximately 25%). However, converting to a change in density by cubing the ratio gives a factor of 2.

One of the signs of many-body ionization is the ionization that occurs for larger separation. The ionization fraction for a pair of ions is less than 1% for separations larger than  $3.5 \times 2n^2a_0$ . However, for more atoms and higher dimensions, there can be substantial ionization for separations larger than this value. In fact, the ionization fraction is approximately 20% for the three dimensional case with  $3^3$  atoms and this separation. This highlights the cooperativity that can occur during ionization.

## B. Randomly Placed Atoms

In this section, we present results when the atoms are randomly placed in a  $d$ -dimensional region. The position of the ions are from a flat random distribution between 0 and  $L$  in each dimension. The separation  $D$  is defined to be  $D = L/N^{1/d}$  where  $N$  is the number of atoms.

In Fig. 4, we plot the fraction of atoms ionized versus the separation of atoms randomly placed on a line. There is an order of magnitude difference in the range of separation compared to Fig. 1. This difference reflects

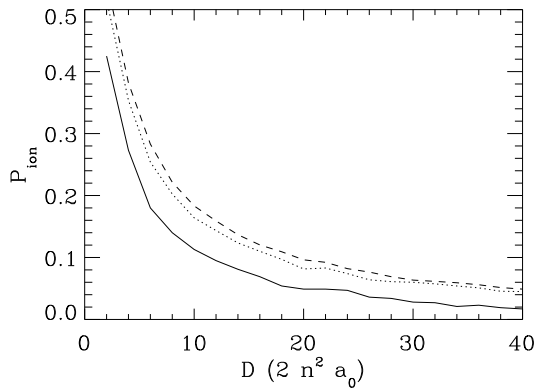


FIG. 4: Same as Fig. 1 but for random placement of atoms. Note the vastly different range for  $D$ .

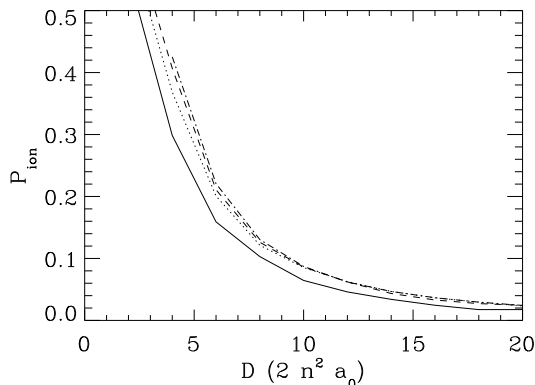


FIG. 5: Same as Fig. 2 but for random placement of atoms. Note the vastly different range for  $D$ .

the qualitative change in ionization when the atoms are randomly placed. Even for large separation, there can randomly be pairs of atoms that are close enough to ionize. If this interpretation is correct, the ionized fraction should be proportional to  $1/D$  for large separation. In fact, the simple function  $2/(D+1)$  is a good approximation to the fraction of ionized atoms for  $D \geq 5$ .

For one dimension, there is an enormous effect from the random placement, but there is also some effect from the many-body interactions. This is reflected in the increase of ionization with number of atoms. For example, there is 10% ionization at  $D \simeq 11$  for 2 atoms, at  $D \simeq 17$  for 4 atoms, and  $D \simeq 19$  for 8 atoms.

We plot the fraction of atoms ionized versus the separation of atoms randomly placed inside a square in Fig. 5. Again, there is a large increase in the range of  $D$  shown compared to Fig. 2. As with the results in Fig. 4, we can attribute this difference to the random placement of atoms and the possibility that random pairs of atoms can be close enough to ionize. If this is the major effect, the ionized fraction should be proportional to  $1/D^2$ . We

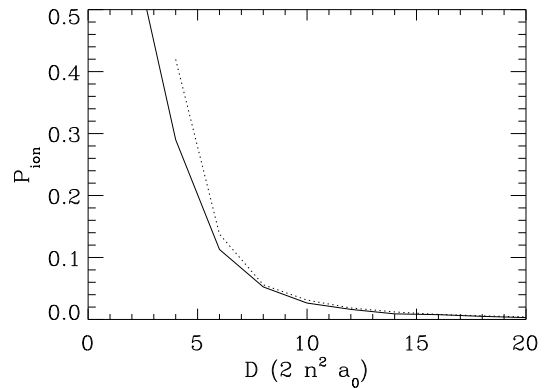


FIG. 6: Same as Fig. 3 but for random placement of atoms. Note the vastly different range for  $D$ .

found that the function  $11/(D+1)^2$  is a good approximation to the fraction of atoms ionized for  $D \geq 5$ .

We plot the fraction of atoms ionized versus the separation of atoms randomly placed inside a cube in Fig. 6. Again, there is a large increase in the range of  $D$  shown compared to Fig. 2. As with the one and two dimensional cases, the density fluctuation appears to be the largest effect. We found that the function  $38/(D+0.6)^3$  is a good approximation to the fraction of atoms that are ionized. There appears to be some effect from many-body ionization but it is difficult to discern when the smaller calculation already has 8 atoms.

To compare the effect of many-body ionization and of fluctuation on the ionization, we can compare the separation where 10% and 20% of the atoms are ionized for a pair of atoms, a cubic grid of atoms and atoms randomly placed in a cube. This discussion is tentative because the cubic grid of atoms does not appear to be converged and, thus, the separation will be underestimated. For 10% ionization, two atoms need a separation of 3.0 compared to 3.8 for the cubic grid and 6.9 for the atoms randomly placed in a cube. Taking this as the measure, the many-body ionization allows a density decrease by a factor of  $(3.8/3.0)^3 \simeq 2.0$  while the fluctuations allow an additional decrease by a factor of  $(6.9/3.8)^3 \simeq 6.0$ . For 20% ionization, two atoms need a separation of 2.8 compared to 3.6 for the cubic grid, and 5.6 for the atoms randomly placed in a cube. Taking this as the measure, the many-body ionization allows a density decrease by a factor of  $(3.6/2.8)^3 \simeq 2.1$  while the fluctuations allow an additional decrease by a factor of  $(5.6/3.6)^3 \simeq 3.8$ . By either measure, the random placement has the larger effect on ionization although the many-body interaction is *not* negligible.

### C. Comparison with experiment

Reference [1] found substantial ionization for densities much smaller than the base density defined in Eq. (1). We can use the results of our calculation for atoms randomly placed in a cube as a comparison. The experiment had different amounts of ionization for somewhat different cases. We will use the density at 10% ionization as our benchmark density; the answer does not qualitatively change if we use a somewhat higher ionization fraction as the benchmark. We obtain a density of

$$\rho_{10\% \text{ ion}} = [(256\pi/3)/(2 \times 6.9)^3]\rho \simeq 0.1\rho. \quad (2)$$

Thus, we obtain substantial ionization for a density an order of magnitude smaller than the base density whereas Ref. [1] had substantial ionization for densities two orders of magnitude smaller than the base density. While an absolute number for the density is hard to obtain experimentally, it seems unlikely to us that the measurement would be wrong by an order of magnitude.

To decrease the density by an order of magnitude, we would need to increase the separation from 6.9 to approximately 14. For this separation we obtain approximately 1% ionization. We have considered two possible mechanisms, not in our calculations, which could increase the fraction of ionized atoms. The first is electron collisions. The  $\sim 1\%$  of promptly ionized electrons could be bound by the space charge effect and then an ionization cascade as in Refs. [3–12] could occur. An argument against this mechanism is that the ionization cascade is typically a slow process compared to the time scales in Ref. [1]. However, the Refs. [3–12] required time to build up the space charge. Perhaps, the prompt ionization in Ref. [1] allows the ionization cascade to start almost instantly. The second mechanism that might be possible is the formation of fast atoms and ions during the ionization step. The calculation of Ref. [2] and the experiment of Ref. [15]

observed that a Penning ionization led to fast Rydberg atoms with a kinetic energy  $\sim 1/5$  of the original binding energy of the cold Rydberg atoms. These fast Rydberg atoms could collide with the much more numerous cold Rydberg atoms causing additional Penning ionization events. A somewhat more complicated variation of this mechanism involves the fast ion undergoing a charge exchange with a cold Rydberg atom which leads to a fast Rydberg atom that can collide with other atoms giving Penning ionization. Performing a realistic simulation of the ionization cascade or the fast Rydberg collisions is beyond the scope of this paper.

### IV. CONCLUSIONS

We have performed classical calculations of prompt ionization in a frozen Rydberg gas. Our calculation fixed the position of the ions but allowed for the full motion of the electrons. The calculations were inspired by the measurements in Ref. [1] which showed substantial ionization for densities two orders of magnitude smaller than a reasonable base density. They attributed the increase in ionization to many-body ionization.

We performed calculations for atoms on a grid and atoms randomly placed within the same volume. By comparing the two calculations, we attribute a factor of  $\sim 2$  increase to many-body ionization and a factor of  $\sim 5$  increase to fluctuation in nearest neighbor separations. We can not account for the extra factor of  $\sim 10$  observed in Ref. [1], but we briefly discussed two possible mechanisms that could increase the ionized fraction of atoms.

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